

Integrated Approach to the Characterization of
Particle and Organic Emissions from
Unvented Kerosene Space Heaters

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KEROSENE SPACE HEATERS

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AN INTEGRATED APPROACH TO THE CHARACTERIZATION OF
PARTICLE AND ORGANIC EMISSIONS FROM UNVENTED
KEROSENE SPACE HEATERS

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Unvented kerosene space heaters (UKSHs) are a major source of indoor air contaminants. This paper reports the results from a phased study designed to characterize and model particle and organic emissions from UKSHs. In Phase I, emissions from 12 heaters covering a range of design types and Btu ratings were evaluated during start-up and three steady state operating conditions (normal, low, and high flame settings). Aerosol mass, size distribution, extractable mass, bioassay, and trace element content, as well as conventional gas (CO , NO_x , etc.) measurements were made for all the heaters tested, while VOCs and semivolatiles measurements were made for a sub-sample of the heaters. In Phase II, a detailed characterization of particle and organic emissions for four UKSHs was performed. Two of the heaters subjected to the detailed chamber testing in Phase II were then used in Phase III. Phase III consisted of a series of heater runs in the U.S. Environmental Protection Agency (EPA) test house in North Carolina. The test house experiments were designed to validate model predictions based on the chamber studies in Phase II.

Introduction

The U.S. Environmental Protection Agency's (EPA) Air and Energy Engineering Research Laboratory (AEERL), Research Triangle Park, North Carolina, has developed an integrated approach to the characterization of indoor air pollutants. The method employs both large and small chambers to characterize pollutant emissions from selected sources. Emission factors developed from these characterization studies are then used to drive an in-house model to predict indoor concentrations of pollutants. The predicted pollutant levels are verified in follow-up testing of the source in the representative environment of AEERL's test house. This approach allows for an interaction between modeling and source measurements to identify and evaluate parameters or phenomena that might have

otherwise been missed. This integrated technique facilitates both source characterization and model validation. More important, this approach allows for predicative capability in other indoor environments.

Unvented kerosene space heaters (UKSHs) were used in the early development of this approach. AEERL and the John B. Pierce Foundation Laboratory, Inc., New Haven, Connecticut, worked together under cooperative agreement during the 1986-1987 heating season to characterize these sources and to develop emissions factors for selected pollutants. The work was implemented in phases. It consisted of a screening phase followed by an intensive characterization in the J. B. Pierce environmental chambers. This was, in turn, followed by test house verification and model validation. This paper presents selected data from the large chamber studies and demonstrates the model by projecting the data into other indoor environments. The actual test house data are contained in another paper presented during this conference¹.

Methods

A total of 10 UKSHs were borrowed from the Consumer Product Safety Commission. These included five radiant (R), one convective (C), and four multistage heaters. The multistage heater consisted of three convective/radiant (C/R) and one multiple radiant (R/R) design. The R/R heater also contained a ceramic catalytic disk mounted above the concentric radiant elements. The test set was further augmented by two C-type heaters obtained locally.

The test group included both new and used heaters of the same type and produced by the same manufacturer. With the exception of a single comparison run, all heaters were fueled with the same 1-K kerosene fuel whose composition had been previously determined by an independent testing laboratory. The heaters were fueled and operated on an "as-received" basis; no effort was made to maltune any of the heaters to obtain "worst case" scenarios.

The UKSH characterization was conducted in the controlled, well-mixed environment of the 34 m³ test chamber located at the J.B. Pierce Foundation Laboratory. Since details of the test protocol have been presented previously, only an overview will be presented here.^{2,3}

The chamber characterization strategy was divided into two phases. Phase I was a general screening step.² In this phase, each UKSH was fueled outside the chamber and subsequently placed in the chamber on a Potter scale which was used to measure fuel consumption. The heater and the chamber were allowed to equilibrate for 1/2 hour, while the recirculating air mixing rate was established and chamber background levels of key parameters were measured. Mixing was maintained with a recirculation rate of 20 air changes per hour (ACH).

The chamber temperature was maintained below 30°C. Cooling was accomplished by recirculating the chamber air over a cooling coil; the temperature was maintained below the dewpoint of the recirculating air to prevent condensation. Fresh air was measured during each run and maintained at approximately 1.2 ACH.

The heater was then fired up and run continuously over a 6-hour period. During the run, the heater was cycled through the range of fuel

consumption/operational settings (i.e., high, low, and normal) that are expected to occur during normal residential use. The actual control settings were determined visually, based on the manufacturer's recommendations that were included with each heater.

Measurements of NO_x , SO_x , CO, CO_2 , and total hydrocarbons were recorded using continuous monitors. The decay of CO_2 at the end of a run was used to calculate the chamber ventilation rate and to provide a measure of reactivity of the other parameters.

Particle size and distribution were measured using an Electrical Aerosol Analyser, a Condensation Nuclei Counter, and an Optical Particle Counter. Integrated mass samples were collected using eight portable Gilian pumps (3 l/min) and an Andersen PM-10 (113 l/min). The latter was modified to accept an XAD-2 cartridge to enable the collection and measurement of semivolatile organics. The particles were collected on tandem Teflon-backed glass fiber filters for gravimetric analysis and bioassay. Volatiles were collected in Summa canisters for GC/MS analysis.

Four heaters were selected from Phase I for further evaluation in an intensive Phase II characterization study. Selection of the four UKSHs was based on criteria intended to pass one heater of each type with the largest particulate mass production to Phase II. The objective of these criteria was to enable the characterization of a set of heaters, any one of which would be capable of generating sufficient collectible particle mass when tested in the test house.

The selected heaters were prepared for testing as in the Phase I screening and the same basic protocol was followed.² The major difference between Phase I and II lay in the sampling interval and the addition of seven more portable pumps for further particle characterization. In Phase II, the UKSHs were operated for 12 hours at a normal flame setting rather than cycled, and replicate experiments were performed. In addition to the previous complement of particle analyses, three pre-fired quartz filters were collected for thermal-optical measurements of organic/elemental carbon by the Desert Research Institute, two PTFE filters were collected for elemental analysis by X-ray fluorescence at NEA, Inc., and two phosphoric-acid-treated quartz filters were collected for inorganic acid analysis at Brookhaven National Laboratory.

At the completion of Phase II, two heaters (one C/R and one R/R) were placed in the test house for further characterization. These heaters were chosen because they represented second generation UKSHs and incorporated burner modifications that were intended to minimize the production of CO. It was believed that these modifications were more representative of future UKSH sales. Details of the test house study are reported separately at this conference.¹

For each chamber run, the steady state pollutant concentration in the chamber was calculated by:

$$G = (\text{Ceq} - \text{Co}) (Q_v + Q_a + Q_r)$$

and $EF = G/F$

where: G = generation rate (mg/hr)

Ceq = measured steady state concentration (mg/hr)

Co = measured ambient/background concentration (mg/hr)
 Qv = chamber ventilation rate (m³/hr) based on the
 measured CO₂ decay rate
 Qa = removal rate by surfaces (m³/hr) estimated from decay
 rates of continuously monitored gases
 Qr = effective removal rate of contaminant by
 collection method being used (m³/hr)
 EF = emission factor (ug/g)
 F = fuel consumption (g/hr)

The derived pollutant concentrations were divided by the fuel consumption on an individual basis to develop emission factors. These emission factors were then modeled using AEERL's Indoor Air Quality (IAQ) model and the results compared to the test house findings. The IAQ model has been described in a previous publication.⁴ It runs on an IBM PC/IBM compatible and is operable via a user-friendly menu-driven interface. The IAQ model can handle up to 10 rooms in a user-defined configuration and can estimate the effect of HVAC systems, outside air exchange, room-to-room air movement, sources, and sinks on indoor air pollutant concentrations.

Results

Taken as a whole, the data indicate that UKSHs are a strong source of indoor respirable suspended particles. Calculated particle emission factors for Phase II ranged from 38 to 376 ug/kJ (see Table I). Particle size distribution measurements indicate that the particles produced by all UKSHs were less than 2.0 um with the peak volume occurring at around 0.15 um.

The largest mass production occurred during start-up. Total mass concentration ranged from 73 to 721 ug/m³. The greatest mass emissions were produced by the R/R heater. Subsequent tests on this specific design indicate that the high levels are probably related to the heater's past-use history.

The aerosol chemical composition data for the UKSHs tested in Phase II are presented in Table II. The sulfur concentration measured as sulfate ranged for 45 to 60 percent of the total particle mass. The apparent form of the sulfate was ammonium sulfate. Significant levels of hydrogen ion concentrations were also measured and will be reported at a later date.

The mass of semivolatile material extracted from the XAD-2 was determined by LC/LLS and by gravimetric analysis and results from the two methods were averaged. The background chamber air contained 34 ug/m³ of extractable material. The concentration of semivolatile materials measured during chamber operation was 98, 24-100, 92, and 379-1192 ug/m³ for the C, C/R, R, and R/R heaters, respectively. The estimated semivolatile emission rates were 11 ± 2, 18 ± 27, 18 ± 5, and 74 ± 62 ug/g fuel for the C, C/R, R, and R/R heaters, respectively.

Volatile organic concentrations as measured by Summa canisters ranged from 20 to 220 ug/m³. A separate aliquot of the unburned 1-K kerosene fuel was analysed by GC/MS and compared to the gases collected in the Summa canisters. The comparison revealed mass spectrographs that

were remarkably similar. The key difference between the two lay in the variations occurring largely in the C12-C15 region. The similarity indicated that volatile components seen in the tests were largely the evaporative emissions from kerosene fuel.

Integrated mass samples collected by the PM-10 were consistently smaller than the mass collected from the portable pumps at the lower flow rate. When backup filters were used in the PM-10, the mass measured from the front filter were on the average 23 percent lower than the levels measured by the single filter on the portable pumps. The combined front and back filter mass averaged 14 percent less than the single filter taken from the portable pump. The difference in masses may be attributable to particle stripping during prolonged sampling and may indicate a limit on the use of PM-10 sampling methods for indoor sources.

Discussion

Initial model runs were made with the emission factors (ug/kJ) developed in the chamber studies and the fuel consumption data from the test house experiments. The air infiltration was assumed to be 0.4 hr^{-1} based on SF_6 measurements in the test house with a window open 2 in. (5 cm). Room-to-room air flows were assumed to be 5 ACH. The measured values were within 2 standard deviations of the modeled values which was acceptable (Figure 1).

The model was then used to predict particle concentrations during an intermittent operating cycle for an indoor environment similar to the test house.¹ The model used the emission factors for the C/R heater (Table I). The heater operating cycle was set for 3 hours in the morning and 6 hours in the evening. The model particle concentrations ranged from approximately 65 to 80 ug/m^3 (Figure 2).

Conversations with EPA's Health Effects Research Laboratory have led us to believe that UKSHs may be a primary heat source for mobile homes.⁵ This fact was substantiated through an extramural project conducted by Applied Management Sciences, Inc. (AMS). The project was designed to survey the population and usage pattern for UKSHs.⁶ AMS estimated that 6.9 million of the estimated 17 million UKSHs sold to date are in use, and that 33 percent of these UKSHs are used in mobile homes.

These housing units, particularly those constructed after 1974 when new standards went into effect, are substantially tighter than standard single family dwellings. It is anticipated that the lower air change coupled with UKSH combustion could lead to higher levels of pollutants in these indoor environments. In addition, AMS stated that UKSHs may be used as a primary heat source due to the reduced area of mobile homes and could be operated as much as 16 hours per day.

Figure 3 depicts modeled particle concentrations for a typical mobile home 14 by 70 ft ($4.3 \times 24.4 \text{ m}$) and 0.25 ACH, using a C/R UKSH (9,916 kJ/hr). The operating cycle is similar to that of the test house in Figure 2. The model shows that, under these conditions, the particle concentrations indoors during operation could be anticipated to range from 50 to 170 ug/m^3 . These predicted concentrations exceed both the 24 hour (150 ug/m^3) and the annual (50 ug/m^3) PM-10 standard. They also compare very well with the 180 ug/m^3 measured in a mobile home known to be using a UKSH as a primary heating source.⁵

Conclusions

The phased approach employing chamber testing, modeling, and test house validation has the important property of being able to predict potential problem areas beyond the scope of limited testing. The test house and mobile home scenarios demonstrate that UKSHs can be a significant source of respirable suspended solids. The modeled concentrations can approximate and, depending on the usage pattern and house characteristics, can exceed the ambient PM-10 standard. Particle levels of this magnitude with a composition similar to that of the UKSH test population provide reason for concern over the potential for adverse health effects arising from the use of UKSHs.

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Table I. Particle emission factors for Phase I/Phase II characterization of unvented kerosene space heaters ug/g (ug/kJ)

Heater Type	Phase I			Phase II		
C	55	6	(1.3)	38		(0.9)
C/R	47	7	(1.1)	42	4	(1.0)
R	58	20	(1.3)	45	1	(1.0)
R/R	218	61	(5.0)	376	47	(8.7)

Phase I emission factors based on cyclic operation over 6 hours.
Phase II emission factors based on normal operation over 12 hours.

Table II. Phase II chemical composition of aerosol

Heater Type	Percent SO ₄	Percent NH ₄
C	60	21
C/R	45	17
R	55	19
R/R	58	24

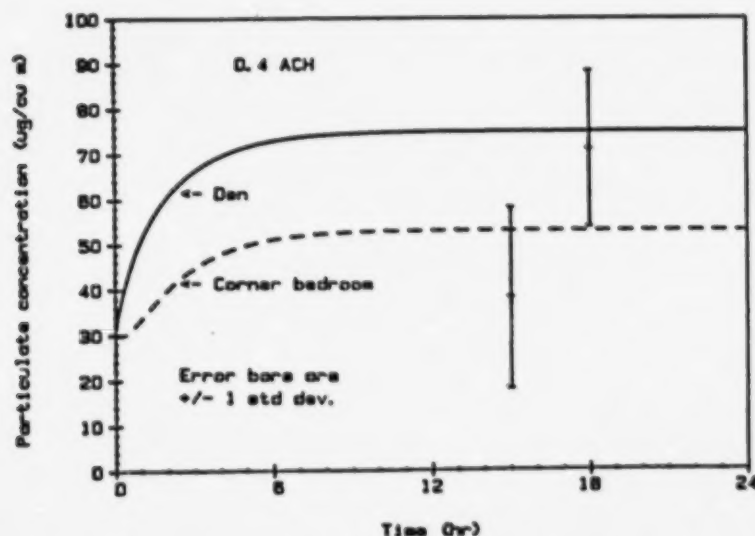


Figure 1. Convective radiant kerosene heater in test house (low room-to-room flow and small sink)

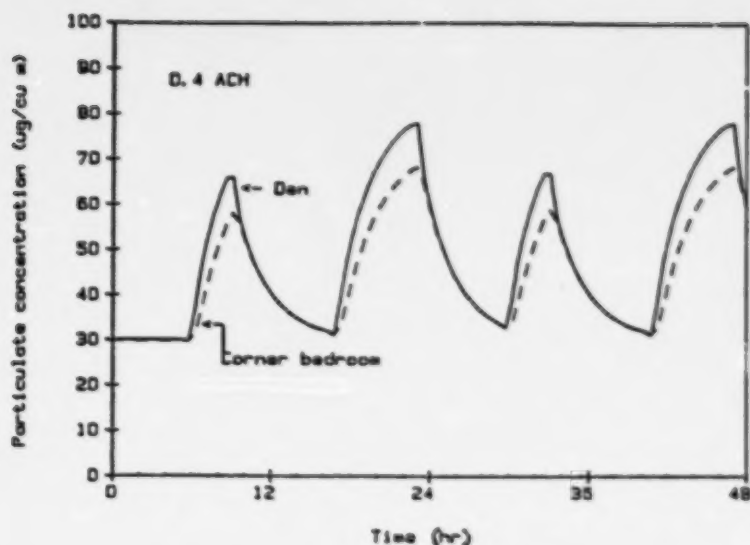


Figure 2. Predicted particulate concentration for convective/radiant heating. (On at 6:00 a.m., off at 9:00 a.m., on at 5:00 p.m., and off at 11:00 p.m.)

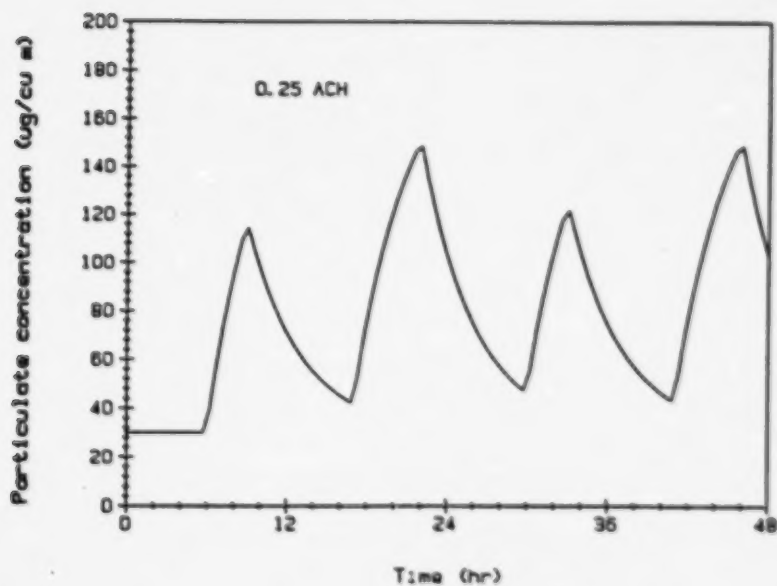


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